

Performance Assessment For Depleted Uranium Disposal In A Near-Surface Disposal Facility

Authors: Karen E. Pinkston, David W. Esh, and Christopher J. Grossman
Affiliation: U.S. Nuclear Regulatory Commission, 11545 Rockville Pike,
Rockville, MD 20852, U.S.A.

ABSTRACT

The U.S. Nuclear Regulatory Commission (NRC) staff has conducted a technical analysis to assess the potential impacts of disposal of large quantities of depleted uranium in a near-surface disposal facility. The nature of the radiological hazards associated with depleted uranium presents challenges to the estimation of long-term effects from its disposal – namely that its radiological hazard gradually increases over time due to the in-growth of decay products. In addition, these decay products include a daughter in gaseous form (Rn-222), which has significantly different mobility in the environment than the parent radionuclides. NRC staff developed a screening performance assessment model of a reference low-level radioactive waste (LLRW) disposal facility to evaluate the risk and uncertainties associated with the disposal of depleted uranium as low-level waste. The model was constructed with the dynamic simulation software package GoldSim®, a Monte Carlo simulation software solution for dynamically modeling complex systems. The depleted uranium source is modeled as releasing to a backfill assumed to surround the depleted uranium in the disposal cells. Radionuclides released to the backfill are vertically transported via advection through unsaturated zone cells to an underlying aquifer, where they are transported to a receptor well. Radon that emanates from radium present in the depleted uranium is modeled as diffusing through an engineered cap into the interior of a residence placed over the disposal area or to the external environment. The model evaluates the radiological risk to future residents and intruders (acute or chronic exposures) near or on the land overlying the disposal facility. Calculations were performed probabilistically to represent the impact of variability and uncertainty on the results. Key variables evaluated included: disposal configurations, performance periods, institutional control periods, wasteforms, site conditions, pathways, and scenarios. The impact of these variables on projected radiological risk can be significant. For example, estimated risks are very sensitive to the performance period, and estimated disposal facility performance is strongly dependent on site specific hydrologic and geochemical conditions. In addition, radon fluxes to the environment are very sensitive to the long-term moisture state of the system and to the disposal depth.

INTRODUCTION

The NRC staff conducted a technical analysis to assess the potential impacts of disposal of large quantities of depleted uranium (DU) in a generic near-surface disposal facility. DU is produced in the enrichment process as a waste product or byproduct. The source term is described as “depleted” because the enrichment process concentrates both the U-235 and U-234 in the product, and therefore, these radionuclides are depleted in the waste or byproduct, which primarily consists of U-238. Staff developed a screening model to evaluate the radiological risk to potential future residents and intruders (acute or chronic exposures) near or on the land

overlying a hypothetical disposal facility for DU and to understand the impacts of key variables on the risks. Key variables evaluated were: disposal configurations, performance periods, institutional control periods, waste forms, site conditions, pathways, and scenarios. The impact of these variables on projected radiological risk can be significant. The model was constructed with the dynamic simulation software package GoldSim®, developed by GoldSim Technology Group of Issaquah, WA, and calculations were performed probabilistically to represent the impact of variability and uncertainty on the results. The model contains 3,252 GoldSim elements, and stochastic inputs are specified for over 400 variables. Conditions such as infiltration rates, liquid saturation, hydraulic gradient, unsaturated zone thickness, hydraulic conductivities, and geochemical conditions were represented in the analysis as epistemic uncertainty to account for a range of sites. In reality, many of these parameters can be constrained for a particular site and disposal system. Because site-specific waste management decisions or other variables can strongly influence whether performance objectives can be met, the results should not be taken out of the analysis context.

THEORY

The nature of the radiological hazards associated with DU presents challenges to the estimation of long-term effects from its disposal because of unique characteristics of the source term. Metallic DU initially contains approximately 99.75 percent U-238, 0.25 percent U-235, and 0.002 percent U-234 [1]. The activity for DU would be expected to remain relatively constant initially, but begin increasing at around 1,000 years as the parent radionuclides decay through the uranium series decay chains. Peak activity, assuming no release from the source, would not be attained until after one million years following disposal. In addition, the activity of some risk significant radionuclides (e.g., Rn-222, Pb-210) increase by a much more significant amount than the overall activity. Because different elements can have different mobility and radiotoxicity, total activity cannot be directly translated to risk (dose). The most prevalent forms of DU for disposal resulting from fuel cycle activities are depleted uranium hexafluoride (UF₆) and depleted uranium oxide (UO₂ or U₃O₈), which results from deconversion of fluoride forms. Both UO₂ and U₃O₈ are solids that are significantly more stable than UF₆ over common disposal conditions, making the oxide forms more suitable for long-term storage or disposal.

DISCUSSION

Evaluation of releases of radioactivity from the disposal of DU was performed for leaching of contaminants to a water pathway and diffusion of radon to the atmosphere. The population was assumed to reside offsite during the institutional control period, and then outside a buffer zone surrounding the disposal area boundary after the institutional control period. The protection of individuals from inadvertent intrusion was evaluated with acute and chronic exposure scenarios following either excavation into the waste, excavation above the waste but not into the waste, or drilling through the waste. The particular intruder scenario evaluated was based on the depth to waste. Below a disposal depth of 3 m (9.8 ft), disruption of the waste via excavation was not believed to be credible for a resident-intruder scenario (see Figure 1). The DU source is modeled as releasing to a backfill assumed to surround the DU in the disposal cells. Radon can partition between the gas and liquid phases, and diffuse in the gas phase through clay, soil, and basement foundation layers, as applicable. Radionuclides released to the backfill are

vertically transported via advection through unsaturated zone cells to an underlying aquifer, where they are transported to a receptor well. Contaminated water is then extracted and used for farming or domestic purposes. The dose was calculated using the probabilistic dose model BDOSETM developed for the NRC by the Center for Nuclear Waste Regulatory Analyses [2]. Exposure pathways in this model include external exposure from surface, air, and water; internal exposure from inhalation of air; and internal exposure from ingestion of drinking water, vegetables/fruits, milk, beef, game, fish, and soil.

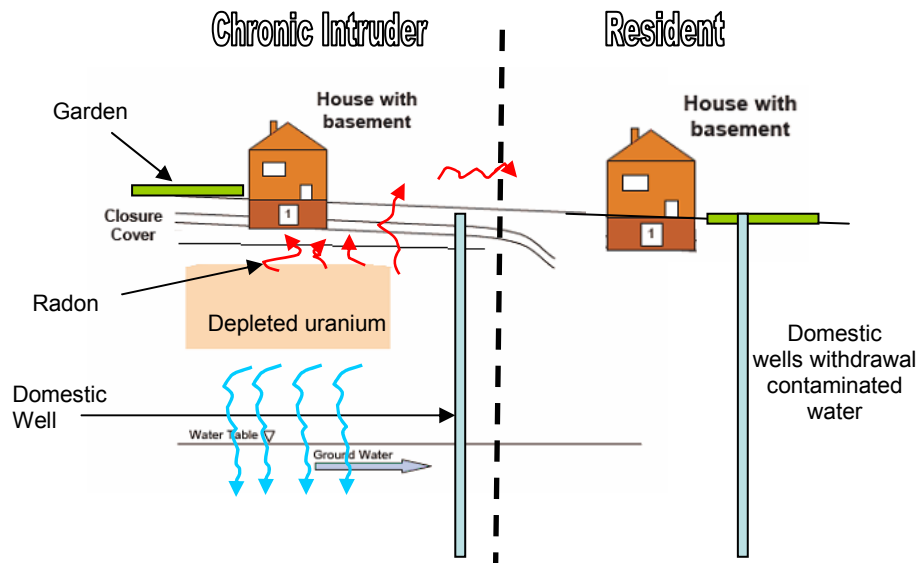


Figure 1. Conceptual Model Showing the Primary Scenarios.

Source term

The source term submodel represents the failure of waste containers over time as well as the gradual degradation of the waste form. The source term submodel applies distribution coefficients, based on material type, to partition radionuclides between solid and liquid phases. Solubility limits are also applied, in addition to partitioning, to estimate liquid phase concentrations of radionuclides. Partition coefficients and solubility values are selected with multi-dimensional lookup tables based on sampled values for pH and carbonate concentration. Numerous references were used to develop the lookup tables [3-8]. It was assumed that depleted uranium would be disposed of in an oxide form. The quantities assumed were 700,000 metric tons (770,000 tons) from DOE and 700,000 metric tons (770,000 tons) from operation of commercial uranium enrichment facilities [9-11]. The disposal system was assumed to have an engineered cover that would limit infiltration. The infiltration cover was assumed to lose its effectiveness a few hundred years after site closure. For arid sites, the long-term infiltration rate was assumed to be on the order of a few millimeters per year. For humid sites the long-term infiltration rate was assumed to be on the order of tens of centimeters per year.

Groundwater transport

The groundwater transport modeling was relatively simple from the perspective of temporal and spatial variability because the assessment was designed to evaluate a range of sites.

Transport through the unsaturated zone was assumed to be vertical to the saturated zone; and transport through the saturated zone was assumed to be horizontal or lateral to a receptors well. Groundwater transport through the unsaturated zone is represented with a series of mixing cells. Advection, partitioning between liquid and solid phases, solubility limits, and decay and in-growth are included in the mathematical representation of a cell. Groundwater transport through the saturated zone is represented with GoldSim pipe elements. Pipes are modeled as reactive columns and include advection, partitioning between liquid and solid phases, decay and in-growth, and dispersion. Because the analysis was generic and hydrologic systems can have widely variable properties, the input distributions were fairly wide, resulting in hydraulic residence times in the pipe from less than ten to greater than 1,000 years.

Radon

Radon that emanates from radium present in the DU is modeled as diffusing to the surface through an engineered cap. The engineered cap contains a clay layer as well as a soil layer. Modeling of radon transport in partially saturated media is subject to a high degree of uncertainty because the gas phase diffusion of radon in partially saturated porous media is highly dependent on the saturation of the media. To take this into account, the tortuosity used in the diffusion calculations is corrected for the saturation of the pore space in the soil and the clay. The outdoor concentration of radon is calculated by modeling the air above the site as a mixing cell in which the radon is diluted and removed by wind. If a residence is located over the DU disposal area, the radon is also modeled as diffusing through the foundation of the house and into the house. Barometric pumping was not included. The validity of this assumption is questionable for shallow disposal depths in arid environments in particular. However, under those conditions, the doses were sufficiently large that the primary output metric of whether the system could meet the performance objectives would not be impacted (i.e., the results already exceeded the performance objectives).

Modeling results

Table I provides the percent of realizations that meet doses of either 0.25 mSv/yr (25 mrem/yr) to the public or 5 mSv/yr (500 mrem/yr) to the intruder for a variety of scenarios and configurations. The results in Table I demonstrate that performance period (i.e., the length of time the analysis was run for), disposal depth at arid sites, and site conditions are important variables to consider for the disposal of DU. Most of the dose at a humid site comes from the groundwater pathway, and the dose at arid sites is mainly due to inhalation of radon. The dose from radon increases greatly at low moisture contents and at shallow disposal depths. With a short performance period, many sites and disposal configurations would be able to meet the performance objectives. For an arid site, radon has not ingrown sufficiently when the performance period is relatively short (e.g., 1,000 years). For both arid and humid sites, the delay in transport may be sufficient to achieve the performance objectives, except for shallow disposal. Disposal of large quantities of DU at depths less than 3 m (9.8 ft) results in projected chronic intruder doses much in excess of 5 mSv/yr (500 mrem/yr). Grouting of the waste may improve the likelihood of an arid site meeting the performance objectives, though the performance of grout over long periods of time is very uncertain.

An uncertainty analysis was performed using genetic variable selection algorithms using a neural network software product, Neuralware NeuralWorks Predict® [12]. For the water dependent pathways at an arid site, important parameters were the hydraulic conductivity and gradient of the aquifer, the infiltration rate, and geochemical conditions that determine sorption and solubilities. For radon at an arid site, the liquid saturation of the materials and properties of the residence and scenario, such as house height, foundation porosity, air exchange rate in the house, and fraction of time spent indoors, were most significant.

Table I. Percent of Probabilistic Realizations that Meet the Performance Objectives

Scenario	Performance Period (yr)	Resident ¹			Chronic Intruder ²
		Total dose	Drinking water	Inhalation	Total dose
Arid, 1 m (3.3 ft) disposal depth	1,000	100	100	100	<2
	10,000	40	90	50	0
	100,000	10	60	20	0
	1,000,000	<1	40	8	0
Arid, 3 m (9.8 ft) disposal depth	1,000	100	100	100	2
	10,000	80	90	100	0
	100,000	50	60	80	0
	1,000,000	20	40	70	0
Arid, 5 m (16 ft) disposal depth	1,000	100	100	100	100
	10,000	80	90	100	100
	100,000	50	60	90	90
	1,000,000	30	40	90	70
Humid, 5 m (16 ft) disposal depth	1,000	70	70	100	100
	10,000	0	0	100	20
	100,000	0	0	100	0
	1,000,000	0	0	97	0
Arid, ³ 5 m (16 ft) disposal depth, Grout	1,000	100	100	100	100
	10,000	90	90	100	100
	100,000	80	80	100	90
	1,000,000	60	60	90	80

¹ Percent of realizations that are below 0.25 mSv/yr (25 mrem/yr) TEDE.

² Percent of realizations that are below 5 mSv/yr (500 mrem/yr) TEDE. When the waste depth is greater than 3 m (9.8 ft), the waste disruption process is through well drilling, not home excavation.

CONCLUSIONS

The model results indicate that the estimated risks from the disposal of DU are sensitive to the performance period and the disposal depth, and estimated disposal facility performance is strongly dependent on site-specific hydrologic and geochemical conditions. Radon fluxes to the environment are very sensitive to the long-term moisture state of the system. Radon is a major contributor to the dose at arid sites with shallow disposal. The groundwater pathway is the major contributor to the dose at humid sites. Grouting of the waste may improve the likelihood of an arid site meeting the performance objectives; however, grout may enhance the mobility of uranium in the groundwater pathway after the grout degrades. It is essential that the site hydrology and geochemistry be well-understood when determining the risk from disposal of large quantities of DU, because site-specific conditions are the primary determinant of the dose. Uranium and daughter radionuclide speciation and partitioning, as well as, radon transport in

natural systems are complex processes; the analysis of the near-surface disposal of DU must adequately evaluate and manage this uncertainty. In particular, measurements of site-specific infiltration rates, radionuclide sorption and solubilities, radon diffusion and emanation rates, waste release rates, and soil-to-plant transfer factors can greatly reduce the uncertainty in the estimated future performance of a disposal site.

REFERENCES

1. Kozak, M.W., T.A Feeney, C.D. Leigh, and H.W. Stockman, 'Performance Assessment of the Proposed Disposal of Depleted Uranium as Class A Low-Level Waste,' Sandia National Laboratories, Albuquerque, NM. 1992.
2. Simpkins, A.A., et al, 'Description of Methodology for Biosphere Dose Model BDOSE.' Center for Nuclear Waste Regulatory Analyses, Southwest Research Institute, San Antonio, TX. 2007.
3. Allard, B., 'Sorption of Cs, I, and Actinides in Concrete Systems.' SKB Technical Report 84-15, Sweden. 1984.
4. Allard, B., 'Chemical Properties of Radionuclides in a Cementitious Environment.' SKB Progress Report 86-09, Sweden. 1987.
5. BSC, 'Dissolved Concentration Limits of Radioactive Elements.' ANL-WIS-MD-000010 Rev 3, Bechtel SAIC Company, Las Vegas, NV. 2004.
6. EPA, 'Understanding Variation in Partition Coefficient, K_d, Values.' EPA-402-R-99-004A. 1999.
7. EPA 'Understanding Variation in Partition Coefficient, K_d, Values. Volume III: Review of Geochemistry and Available K_d Values for Americium, Arsenic, Curium, Iodine, Neptunium, Radium, and Technetium' EPA-402-R-04-002C. 2004.
8. Sheppard, M.I. and D.H. Thibault. *Default Soil Solid/Liquid Partition Coefficients, K_ds, for Four Major Soil Types: A Compendium. Health Physics.* Vol. 59. pp. 471–482. 1990.
9. U.S. Department of Energy (DOE). 'Draft Supplement Analysis for Location(s) to Dispose of Depleted Uranium Oxide Conversion Product Generated from DOE's Inventory of Depleted Uranium Hexafluoride.' DOE/EIS-0359-SA1. Office of Environmental Management. 2007.
10. NRC, 'Environmental Impact Statement for the Proposed National Enrichment Facility in Lea County, New Mexico, Final Report.' NUREG-1790, June 2005.
11. NRC, 'Environmental Impact Statement for the Proposed American Centrifuge Plant in Piketon, Ohio, Final Report.' NUREG-1834, 2006.
12. Neuralware, NeuralWorks Predict® Product Version 2.40, Carnegie, PA. 2001.